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PATENT SPECIFICATION

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- (21) Application No. 25113/70 (22) Filed 26 May 1970
(31) Convention Application No. 828337 (32) Filed 27 May 1969
(31) Convention Application No. 839648 (32) Filed 7 July 1969 in
(33) United States of America (US)



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The following corrections were allowed under Section 76 on 5 September 1972:-

Page 3, line 55, *for trimelletic read trimellitic*

Page 3, line 84, *after pounds insert such*

Page 5, line 8, *for alkyol read alkylol*

Page 5, line 117, *for and read an*

Page 8, line 5, *for ats. read at*

Attention is also directed to the following printers' errors:-

Page 1, line 1, *for PpG read PPG*

Page 2, line 7, *for variouus read various*

Page 2, line 9, *for Reistance read Resistance*

Page 2, line 12, *for humdity read humidity*

Page 2, line 15, *for coating read coated*

Page 2, line 54, *for poyether read polyether*

Page 2, line 76, *for polyester read polyether*

Page 2, line 86, *after coatings delete full stop*

Page 2, line 108, *for of read or*

Page 3, line 27, *for trimethylolalethane read trimethylolalethane*

Page 4, line 102, *for tmployed read employed*

Page 5, line 60, *for properties read proportions*

Page 5, line 73, Page 9, line 57, *delete the*

Page 8, line 30, *for equimolar read equivalent*

Page 8, line 45, *for methane-cis read methane-bis*

Page 9, line 8, *after polyether insert polyol*

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(54) COATING COMPOSITION

- (71) We, PPG INDUSTRIES, INC., a Corporation organized under the laws of the State of Pennsylvania, United States of America, of One Gateway Center, Pittsburgh, State of Pennsylvania, United States of America, (assignee of WEN-HSUAN CHANG and MARVIS EDGAR HARTMAN), do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—
- The present invention relates to a coating composition.
- Recent advances in coating technology have provided coatings which are suitable for use over various substrates which are difficult to coat and having many different properties. Coatings of excellent appearance, a high order of durability and having the ability to withstand severe environmental conditions have been obtained. Among the more advanced coatings are those employed on vehicles, such as automobiles, where good appearance must be maintained over long periods despite exposure to weather and various forms of attack during use.
- Recently, there has been interest in the use of rubbery, resilient materials for areas which are subject to mechanical shock, such as automobile bumpers and moldings, exposed corners and surfaces of industrial machines, kickplates and other areas of doors and entrances. The use of such materials aids in providing protection against permanent structural damage, but in order to attain the desired appearance a decorative and protective coating must be applied to the surface and this coating can also be damaged during use. Conventional coatings, including those employed on rubber and similar extensible objects heretofore, do not have the required combination of properties. These necessary properties include:
1. Extensibility—This property is necessary in order that the advantages of the resilient substrate can be utilized without destruction of the integrity of the surface of the coating.
 2. Tensile Strength—A high degree of tensile strength is also necessary in order to avoid rupture of the film during use.
 3. Package Stability—In order to permit ease of application, the liquid coating composition should be stable for extended periods under ambient conditions without either gelation or depolymerization of the resin contained therein.
 4. Film Stability—Certain coatings which are extensible and which have a relatively high tensile strength lose these properties upon aging and particularly if the coating is exposed to sunlight, weathering, etc.
 5. Impact Resistance—The coating should have adequate impact resistance at various temperatures as encountered in extreme weather variations, including temperatures as low as -20°F . and as high as 120°F .
 6. Adhesion—The coating should have satisfactory adhesion to the various substrates with which it is to be employed including extensible materials such as foams and rubber, and metals such as mild steel. In addition the

SEE CORRECTION SLIP ATTACHED

coatings should have satisfactory intercoat adhesion with succeeding coats or with various primers which can be employed.

- 5 7. Chemical and Humidity Resistance—This includes properties such as saponification resistance upon exposure to acids and alkalis, resistance to various solvents and resistance to atmospheres of high humidity and heat.

- 10 8. Resistance to Cracking under Temperature-Humidity cycling—This property is important where the coating might be exposed to rapid variations in temperature and humidity as might be encountered by automobiles during travel or storage. This property is tested
15 by successively exposing the coating object to conditions of high temperature and high humidity alternated with exposure to low temperature and low humidity.

- 20 Still other properties which are important for commercial applicability include sprayability at reasonable solids contents; a non-toxicity, and low sensitivity to moisture.

- 25 It is especially difficult to obtain the above properties in combination since, in most instances, the obtention of one or several of the properties desired requires the use of materials and formulations which under ordinary circumstances tend to make the other desired properties less satisfactory.

- 30 According to the present invention there is provided a one-package, storage-stable coating composition comprising

- 35 (A) an ungelled, hydroxyl-containing urethane reaction product of an organic polyisocyanate and a polyhydric material comprising at least 50% by weight of a component selected from

- (1) a polyester polyol formed from
40 (a) an alcohol component having an average functionality of at least 1.9;
(b) an acid component comprising one or more monomeric carboxylic acids or anhydrides having 2 to 14 carbon atoms per molecule, said acid component having an average functionality
45 of at least 1.9;
said alcohol component and said acid component containing a total of not more than one gram-mole of compounds having a functionality of 3 or more per 500 grams of (a) plus (b); and
50 (2) a polyhydric material comprising a major proportion of a polyether polyol having a hydroxyl equivalent of at least 100 and being formed from a total of not more than one gram-mole of compounds having a functionality of 3 or more per 500 grams of polyether polyol and a minor proportion of a diol or a polyester polyol; and
55 (B) an amino plast resin.
60

The polyester is formed from an alcohol

component having an average functionality of at least 1.9 and an acid component comprising one or more monomeric carboxylic acids or anhydrides containing 2 to 14 carbon atoms and also having an average functionality of at least 1.9. To obtain the desired extensibility and other properties, the alcohols and acids or anhydrides used should contain a total of not more than one gram-mole of compounds having a functionality of 3 or more per 500 grams of the total of the alcohols and acid or anhydride employed.

The polyester polyol has a hydroxyl equivalent of at least 100, and usually between 100 and 10,000. (By "hydroxyl equivalent" is meant the weight per hydroxyl group). To obtain the desired extensibility and other properties, the polyether polyol should be produced from a total of not more than one gram-mole of compounds having a functionality of 3 or more per 500 grams of the total of the components.

Such coatings can be applied to virtually any solid substrate and are especially useful on rubbery, resilient substrates such as polyurethane or polyethylene foam, natural or synthetic rubber or rubber foam, and various elastomeric plastic materials. They are also particularly useful on either substrates such as mild steel or aluminium.

These compositions, although based on a urethane system, are distinguished from ordinary polyurethanes because they are both storage-stable in one package and yet form a cross-linked thermoset cured coating.

The coatings herein provide the above-mentioned properties to a satisfactory degree and have a combination of these properties which is not obtainable with conventional coating systems.

The compositions of the invention contain as one component an isocyanate-modified resin containing hydroxyl groups and formed by reacting a polyhydric material comprising either a substantially linear polyester polyol of a polyether polyol with an organic polyisocyanate. The isocyanate-modified resin is combined with an aminoplast resin to provide the coating composition of the invention.

It is necessary that the polyester polyol or polyether polyol employed have certain properties in order to provide a coating of the desired characteristics. These properties are obtained, in general, by utilizing either a polyester derived from a polyol having an average functionality of at least 1.9 or a polyether polyol (or a mixture of polyols) having relatively long chains per hydroxyl group and which thus has a hydroxyl equivalent of at least 100 and preferably at least 300. The polyol component in most cases consists essentially of one or more diols; triols, or higher polyols can also be used in whole or in part provided the polyether polyol component is not derived from more than 1 gram mole of

The polyester is produced using conventional techniques, with the reaction conditions and the ratio of reactants chosen so as to provide a product having residual hydroxyl groups. The number of hydroxyls present in the product can be varied, but it is preferred that its hydroxyl value be at least 30 and preferably more than 80.

When polyether polyols are used as the polyhydric material in this invention they are used with minor amounts of diols, such as 1,4-butanediol and neopentyl glycol or polyester polyols. There can also be included monohydric alcohols; and polyfunctional compounds containing one or more hydroxyls, such as ethanolamine and compounds comprising other active hydrogen-containing groups, such as water and polyfunctional amines; examples include isophorone diamine, p-menthane diamine, propylene diamine, hexamethylene diamine, diethylene triamine, triethylene tetramine and diethanolamine.

The organic polyisocyanate which is reacted with the polyhydric material as described is essentially any polyisocyanate, e.g., hydrocarbon polyisocyanates or substituted hydrocarbon diisocyanates. Many such organic polyisocyanates are known in the art, including p-phenylene diisocyanate, biphenyl diisocyanates, toluene diisocyanates, 3,3'-dimethyl-4,4'-biphenylene diisocyanate, 1,4-tetramethylene diisocyanate, hexamethylene diisocyanate, 2,2,4-trimethyl-1,6-hexylene diisocyanate, methylene bis(phenyl isocyanate), lysine methyl ester diisocyanate, bis(isocyanatoethyl) fumarate, isophorone diisocyanate and methyl cyclohexyl diisocyanates. There can also be employed isocyanato-terminated adducts of diols, such as ethylene glycol, 1,4-butylene glycol and polyalkylene glycols. These are formed by reacting more than one mole of a diisocyanate, such as those mentioned, with one mole of a diol to form a longer chain diisocyanate. Alternatively, the diol can be added along with the diisocyanate.

While diisocyanates are preferred, higher polyisocyanates can be utilized as part of the organic polyisocyanate. Examples are 1,2,4-benzene triisocyanate and polymethylene polyphenyl isocyanate.

It is preferred to employ an aliphatic diisocyanate, since it has been found that these provide better color stability in the finished coating. Examples include bis(isocyanatocyclohexyl)methane, 1,4-butylene diisocyanate and methylcyclohexyl diisocyanates. The proportions of the diisocyanate and the polyether polyol are chosen so as to provide a hydroxyl-containing product. This can be accomplished by utilizing a less than stoichiometric amount of polyisocyanate, i.e., less than one isocyanate group per hydroxyl and carboxyl group in the polyether. Higher (e.g., stoichiometric or excess) isocyanate levels can be present if the reaction is terminated at the

desired stage, as by addition of a compound which reacts with the residual isocyanate groups; water, alcohols and amines are examples of such compounds.

In one especially desirable embodiment of the invention, a polyfunctional alcohol is used to terminate the reaction at the desired state (determined by the viscosity), thereby also contributing residual hydroxyl groups. Particularly desirable for such purposes are aminoalcohols, such as ethanolamine and diethanolamine, since the amino groups preferentially react with the isocyanate groups present. Polyols, such as ethylene glycol, trimethylolpropane and hydroxyl-terminated polyesters or polyethers, can also be employed in this manner.

While the ratios of the components of the polyester polyol or the polyether polyol, polyisocyanate, and any terminating or blocking agent can be varied, it will be noted by those skilled in the art that the amounts should be chosen so as to avoid gelation and to produce an ungelled, urethane reaction product containing hydroxyl groups. The hydroxyl value of the urethane reaction product should be at least 10 and preferably 20 to 200.

The urethane reaction product as described above is mixed with an aminoplast resin to provide the coating composition. Aminoplast resins are aldehyde condensation products of melamine, urea, and similar compounds; products obtained from the reaction of formaldehyde with melamine, urea or benzoguanamine are most common and are preferred herein. However, condensation products of other amines and amides can also be employed, for example, aldehyde condensates of triazines, diazines, triazoles, guanidines, guanamines and alkyl and aryl substituted derivatives of such compounds, including alkyl and aryl substituted ureas and alkyl and aryl substituted melamines. Some examples of such compounds are N,N-dimethylurea, benzourea, dicyandiamide, formoguanamine, acetoguanamine, ammeline, 2-chloro-4,6-diamino-1,3,5-triazine, 6-methyl-2,4-diamino-1,3,5-triazine, 3,5-diaminotriazole, triaminopyrimidine, 2-mercapto-4,6-diaminopyrimidine and 2,4,6-triethyl triamino-1,3,5-triazine.

While the aldehyde employed is most often formaldehyde, other similar condensation products can be made from other aldehydes, such as acetaldehyde, crotonaldehyde, acrolein, benzaldehyde, furfural, and others.

The amine-aldehyde condensation products contain methylol or similar alkylol groups, and in most instances at least a portion of these alkylol groups are etherified by a reaction with an alcohol to provide organic solvent-soluble resins. Any monohydric alcohol can be employed for this purpose, including such alcohols as methanol, ethanol, propanol, butanol, pentanol, hexanol, and heptanol, as well as benzyl alcohol and other aromatic

alcohols, cycloaliphatic alcohols such as cyclohexanol, monoethers of glycols such as Cellosolves and Carbitols (CELLOSOLVE and CARBITOL are Trade Marks), and halogen-substituted or other substituted alcohols, such as 3-chloropropanol. The preferred amine-aldehyde resins are etherified with methanol or butanol. In some instances, the alkyl groups of the condensation product are reacted to form acyl groups, as by reaction with acetic anhydride, or are reacted with amines such as morpholine.

For optimum properties, it is preferred that the composition contain a polymeric polyol having a low glass transition temperature, i.e., having a glass transition temperature below 25°C. The inclusion of such a polymeric polyol gives a balance of flexibility and hardness. In many cases the polyether polyol utilized in the formation of the urethane product has the desired glass transition temperature. When it is desired to add a component of this type, there can be used in addition to or in lieu of the polyether polyols described above, any of the many polyester polyols having the desired glass transition temperature, especially those produced from acyclic reactants such as adipic acid and azelaic acid and alkylene glycols; poly (neopentyl adipate) is a useful example. Still other polymeric polyols of suitable properties include condensates of lactones with polyols, such as the product from caprolactone and ethylene glycol; propylene glycol or trimethylolpropane.

Where the polyether polyol utilized in preparing the urethane reaction product is of low glass transition temperature, it is often found that the coating does not have suitable hardness. It is therefore desirable in many cases to employ the "soft" polyether polyol in conjunction with a minor proportion of a polyester polyol, or the constituents thereof, having a higher glass transition temperature. One method is to include polyester moieties in the polyether molecule; another way is to produce an isocyanato-terminated adduct or prepolymer from the polyester polyol and the polyisocyanate and then react the prepolymer with the polyether polyol; a third method is to blend the polyester polyol as such with the polyether polyol before or after the polyester is reacted with the polyisocyanate. Low molecular weight diols or diamines can also be employed in conjunction with the polyether polyol. The choice of method depends upon the particular components used and the properties desired, but in each instance the product obtained contains both "hard" and "soft" segments in a type of block copolymer.

The properties of the above components can be varied to provide certain properties. For example, higher levels of polyether polyol in the urethane reaction product result in somewhat softer and more extensible coatings, whereas harder, more resistant coatings are

obtained by increasing the proportion of aminoplast resin. The amounts employed depend in large part upon the nature of the particular components, e.g., the specific polyether polyol, aminoplast resin, as well as the type of polyester polyol, if any, employed. In most cases the overall composition contains from 50 to 95 percent by the weight of urethane reaction product, and from 5 to 50 percent of aminoplast resin.

The compositions herein can be applied by any conventional method, including brushing, dipping and flow coating, but they are most often applied by spraying. Usual spray techniques and equipment are utilized. They can be applied over virtually any substrate, including wood, metals, glass, cloth, plastics and foams, as well as over various primers.

The coatings are cured at elevated temperatures. In most cases the cure schedule is from 20 to 40 minutes at 140°F. to 260°F. Higher or lower temperatures with correspondingly shorter and lower times can be utilized, although the exact cure schedule best employed depends in part upon the nature of the substrate as well as the particular components of the composition. Acid catalysts and other curing catalysts can be added to aid in curing if desired; these can permit the use of lower temperatures and/or shorter times.

The invention will be further described in connection with several examples which follow. These examples are given as illustrative of the invention and are not to be construed as limiting it to their details. All parts and percentages in the examples and throughout the specification are by weight unless otherwise indicated.

EXAMPLE 1

The following were charged to a reaction vessel:

	Parts by Weight	
Neopentyl glycol	126.9	110
Trimethylolpropane	22.1	
Adipic acid	72.3	
Isophthalic acid	123.2	

This mixture was heated to 200°C. for 30 minutes and then at 220°C. until the resin had a Gardner-Holdt viscosity of F (60 percent solids in methyl ethyl ketone), and acid value of about 10 and a hydroxyl value of about 100. This polyester polyol was then mixed with the following:

	Parts by Weight	
Polyester	70	
Methyl ethyl ketone	35	
Methane-bis(cyclohexyl isocyanate)*	7.13	125

* Mobay D-244

This mixture was heated at 150°F. for 20 hours and then cooled to 120°F. for 3 more hours. There were then added 22 parts of n-butanol and 0.3 part of ethanolamine. The product has a Gardner-Holdt viscosity of Z1—Z2, a non-volatile solids content of about 60 percent and an acid value of 3.7.

A gray coating composition was formulated using the urethane reaction product thus produced by blending the following:

	Parts by Weight
Urethane reaction product	196.5
Hexakis (methoxymethyl)melamine ("Cymel 301")	18.7
Poly(oxytetramethylene) glycol	15.8
CAB solution*	7.5
Pigment paste	67.5
Toluene	217.5
Cellosolve acetate	49.5
Ethyl Cellosolve	99
Diacetone alcohol	74.5
p-Toluene sulfonic acid	1.5
* 20 percent solution in $\frac{1}{2}$ second cellulose acetate butyrate in 80/20 toluene/ethanol.	

The pigment paste employed was ground in a solution of a polyester made from 146 parts of neopentyl glycol, 112 parts of adipic acid, 191 parts of isophthalic acid and 103 parts of trimethylolpropane; the paste was produced by mixing the following:

	Parts by Weight
Polyester (60 percent solids in xylene)	89
TiO ₂	260
Molacco black	13
Xylene	28
Methyl isobutyl ketone	43.5
Butanol	10.5

This mixture was ground in a ball mill until the particles had a fineness of 6—1/2 Hegman and then 50 parts of toluene were added.

The coating composition thus obtained had good storage stability and excellent properties. For example, when baked at 250°F. for 60 minutes it provided a film having outstanding extensibility (maximum elongation 181 percent) and tensile strength (2.09×10^3 g/cm², measured on Instron Tester—INSTRON is a Trade Mark). When coated on polyurethane foam of the type employed for automobile bumpers (having a density of 40 pounds per cubic foot and a Shore "A" hardness of 78), it had desirable properties, including impact resistance in excess of 160 inch/lbs (measured on a Gardner Variable Impact Tester). This composition is suitable as a primer over such foams and similar materials.

EXAMPLE 2

A white coating composition was produced using the urethane reaction product described in Example 1 and a pigment paste made in a manner similar to that described in Example 1 but without carbon black; the pigment contained 61.5 percent TiO₂ and 12 percent of polyester along with suitable solvents. The coating composition had the following constituents:

	Parts by Weight	
Urethane reaction product	74	
Hexakis(methoxymethyl)melamine ("Cymel 301")	10	
Pigment paste	40	
Poly(oxytetramethylene glycol) (mol. wt. 1000)	6	
CAB solution (as in Example 1)	5	
p-Toluene sulfonic acid	0.5	
Methyl ethyl ketone	100	
Cellosolve acetate	50	

Films of this composition (baked 60 minutes at 250°F.) had a maximum elongation of 177 percent and tensile strength of 1.55×10^3 g/cm². When coated over the primed foam produced in Example 1, it provided a coated product of outstanding properties including good impact resistance.

EXAMPLE 3

A coating composition was produced as in Example 1 except that the aminoplast resin utilized was a more resinous, less highly methylated melamine-formaldehyde resin known as "QR-483". The product had properties substantially similar to that from Example 1.

EXAMPLE 4

Following the procedure of Example 1, a coating composition was produced using the urethane reaction product and pigment paste described therein, but with a butylated melamine-formaldehyde resin made using 5.5 moles of formaldehyde and 6 moles of butanol per mole of melamine; the resin was a 60 percent solids solution in 25/75 butanol/xylene. The coating composition contained the following:

	Parts by Weight	
Urethane reaction product	110	
Butylated melamine resin	42	
Poly(oxytetramethylene)glycol (mol. wt. 1000)	10.5	
Pigment paste	45	
CAB solution (as in Example 1)	5	
Xylene	130	
Cellosolve acetate	40	
Ethyl Cellosolve	80	
Diacetone alcohol	50	
p-Toluene sulfonic acid	2	

- good storage stability and excellent properties; when applied over poly (vinyl chloride) foam and baked at 250°F. for 30 minutes it provides a coating having good low temperature impact resistance (3 ft.-lbs. ats.—20°F.), and tensile strength (2.5×10^3 g/cm², measured on Instron Tester).

EXAMPLE 7

- Example 6 is repeated except that the aminoplast resin employed is hexakis(methoxymethyl)melamine ("Cymel 300"). Substantially similar properties are obtained.

EXAMPLE 8

- Example 6 is repeated using as the aminoplast resin a butylated melamine-formaldehyde resin made using 5.5 moles of formaldehyde and 6 moles of butanol per mole of melamine. Good results are obtained.

EXAMPLE 9

- Example 6 is repeated using a urethane reaction product formed from the following:

	Parts by Weight
Poly(oxytetramethylene)glycol (mol. wt. 1000)	352
1,4-Butanediol	34
1,6-Hexanediol	45
Polyester polyol (as in Example 1)	202
Methane-bis (cyclohexyl isocyanate)	367

- Substantially equimolar results to those of Example 6 are obtained.

EXAMPLE 10

- A urethane reaction product, employed in a manner similar to the above examples, is produced as follows:

	Parts by Weight
Poly(oxytetramethylene)glycol (mol. wt. 1000)	500
1,4-Butanediol	105
1,6-Hexanediol	150
Trimethylolpropane	30
Methyl isobutyl ketone	1835

- The above mixture (1000 parts) is mixed with 292 parts of methane-cis(cyclohexyl isocyanate), 680 parts of methyl isobutyl ketone and 2 parts of a 1 percent solution of dibutyl tin diacetate in methyl isobutyl ketone, and heated at 230°C. for 10 hours. There are then added 30 parts of butanol per hundred parts of resin.

EXAMPLE 11

- A urethane reaction product is produced in a manner similar to that described in Example 10, from the following:

	Parts by Weight	
Poly(oxytetramethylene)glycol	500	
Isophorone diamine	10.5	
Diethanolamine	3	60
1,4-Butanediol	85	
Toluene diisocyanate*	300	
* 80 percent, 2,4-isomer, 20 percent 2,6-isomer		

- Employed in the manner of the above examples, coatings of suitable properties are obtained.

In a similar manner, coating compositions of desirable properties are produced using other polyether polyols of the class described and other polyester polyols, as well as other polyisocyanates in place of the polyethers and polyisocyanates in the examples. Also, while the examples show the inclusion of certain diols, other polyols can be used instead; such polyols include, for instance, monomeric diols and triols.

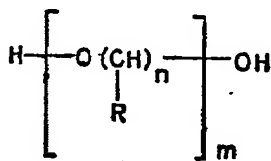
WHAT WE CLAIM IS:—

1. A one-package, storage-stable coating composition comprising
 - (A) an ungelled, hydroxyl-containing urethane reaction product of an organic polyisocyanate and a polyhydric material comprising at least 50% by weight of a component selected from
 - (1) a polyester polyol formed from
 - (a) an alcohol component having an average functionality of at least 1.9;
 - (b) an acid component comprising one or more monomeric carboxylic acids or anhydrides having 2 to 14 carbon atoms per molecule, said acid component having an average functionality of at least 1.9;
 - said alcohol component and said acid component containing a total of not more than one gram-mole of compounds having a functionality of 3 or more per 500 grams of (a) plus (b); and
 - (2) a polyhydric material comprising a major proportion of a polyether polyol having a hydroxyl equivalent of at least 100 and being formed from a total of not more than one gram-mole of compounds having a functionality of 3 or more per 500 grams of polyether polyol and a minor proportion of a diol or a polyester polyol; and
 - (B) an amino plast resin.
2. A composition as claimed in claim 1 in which the acid component of the polyester polyol contains at least 75 mole percent of dicarboxylic acids or anhydrides.
3. A composition as claimed in claim 1 or 2 in which the polyester polyol has a hydroxyl value of at least 30.
4. A composition as claimed in claim 3 in

which the polyester polyol has a hydroxyl value of more than 80.

- 5 A composition as claimed in any preceding claim in which the polyester polyol is the reaction product of adipic acid and neopentyl glycol.

6. A composition as claimed in claim 1 in which the polyether has the general formula



- 10 wherein R is a hydrogen or lower alkyl having up to 6 carbon atoms, n is from 2 to 6 and m is from 2 to 100.

7. A composition as claimed in claim 6 in which the polyether glycol is a poly (oxytetramethylene) glycol.

8. A composition as claimed in claim 7 in which the poly (oxytetramethylene) glycol has a molecular weight between 400 and 10,000.

9. A composition as claimed in claim 1 in which the polyether polyol is one formed by reacting a polyol with an alkylene oxide in the presence of an acidic or basic catalyst.

10. A composition as claimed in any preceding claim in which the polyether polyol has a hydroxyl equivalent not above 10,000.

11. A composition as claimed in any preceding claim in which the polyhydric material also comprises other hydroxyl-containing compounds and/or compounds containing other active-hydrogen-containing groups.

12. A composition as claimed in any preceding claim in which the polymeric polyol has a glass transition temperature below 25°C.

13. A composition as claimed in any preceding claim in which the polyisocyanate is a diisocyanate.

14. A composition as claimed in claim 13 in which the diisocyanate is an aliphatic diisocyanate.

15. A composition as claimed in any preceding claim in which the hydroxyl value of the urethane reaction product is at least 10.

16. A composition as claimed in claim 15 in which the hydroxyl value of the urethane reaction product is from 20 to 200.

17. A composition as claimed in any preceding claim in which the aminoplast resin is an alkylated condensation product of formaldehyde and melamine, urea or benzoguanamine.

18. A composition as claimed in any preceding claim which contains from 50 to 95 percent by weight of urethane reaction product and from 5 to 50 percent by weight of aminoplast resin.

19. A composition as claimed in any preceding claim in which the polymeric polyol is blended with the urethane reaction product and aminoplast resin.

20. A composition as claimed in claim 1 substantially as hereinbefore described with reference to any of the foregoing Examples.

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Chartered Patent Agents.

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